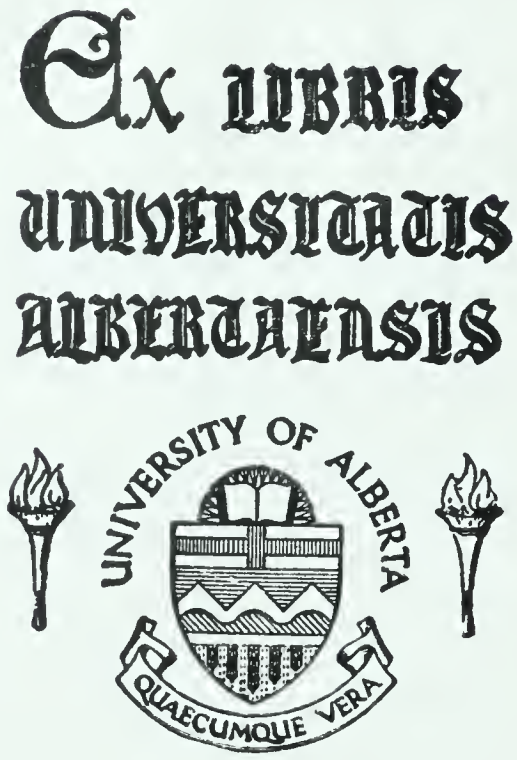

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GAS SCINTILLATION

A THESIS

SUBMITTED TO THE FACULTY OF GRADUATE STUDIES IN

PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF

MASTER OF SCIENCE

DEPARTMENT OF PHYSICS

BY

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UNIVERSITY OF ALBERTA

FACULTY OF GRADUATE STUDIES

The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies for acceptance, a thesis entitled Gas Scintillation, submitted by J.F. Easton in partial fulfilment of the requirements for the degree of Master of Science.

ABSTRACT

This thesis describes the building of a gas scintillator and experiments done to measure the effects of impurities.

It is concluded that a practical neutron counter with a stable lifetime in excess of one week can be built and recommendations are given for the building of such a counter.

It is concluded that for low energy neutrons the counter would be useful for giving a timing pulse in time of flight work and would itself measure the energy of higher energy neutrons (1 Mev.) with a reasonable resolution.

ACKNOWLEDGEMENTS

I would like to thank:

Dr. G. C. Neilson for suggesting the project.

Dr. R. H. Krouse for doing the mass spectrometer analysis of the gas.

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I INTRODUCTION

The passage of charged particles through a material causes the atoms in the material to emit radiation. This process, known as scintillation, has been used for many years in the detection of charged particles.

Of interest in the present work is the scintillation of the inert gases xenon, krypton, argon and helium. Because of the possibility of using the $He^3(n,p)H^3$ reaction (ref. 2 & 3) and the $Ne^{20}(n,a)O^{17}$ reaction (ref. 4 & 6) as neutron detection devices, it is of interest to investigate the scintillating properties of mixtures of these gases.

The first successful observation of gas scintillations was by C.O. Muehlhouse in 1953 (ref. 8), who observed a barely detectable light output from argon and helium using Pu alpha particles. Since then, much work has been done and considerable improvements have been made. It has been established that pure gas counters have two distinct advantages; they are insensitive to γ-rays (down by a factor of at least 20 (ref.3)), and the response time is short (less than 10 nsec. (ref. 2)). The first successful use of gas counters was in an experiment undertaken by R.A.Nobles, R.L. Henkle and P.K. Smith (ref. 9), who used a xenon counter to measure the neutron fission cross-section of Am^{241}. This nucleus has a high specific activity and the fast recovery time of xenon enabled them to overcome the pileup of pulses due to the alpha particles.

It was intended that a counter using xenon and He^3 as mentioned above be constructed for use as a neutron detector. The Q of the reaction and the high cross-section for low energy neutrons would make such a counter useful for time of flight work at low energies. It has been found by other workers that gas purity is of utmost importance and has proved to be the most serious problem encountered.

Since there is some disagreement in the literature regarding the

effects of nitrogen in such a counter it was thought that the effects of low level impurities should be investigated. A counter and a calcium purifier are described with the results of impurity measurements. Enough is now known about gas counters to build a neutron counter and the behaviour of such a counter can be predicted with a fair degree of accuracy.

II Survey of past work

There are some properties of gas scintillation counters about which there is agreement in the literature. The first, and perhaps, basically the most important fact, is that the light emitted by the monoatomic gases falls predominantly in the ultra-violet region. Measured wavelengths as far down as 1600 A have been reported. (ref. 4) Using a photomultiplier tube with a quartz face one can expect to improve upon the conditions one would have using an ordinary photomultiplier. Quartz window tubes (RCA 6903) have a maximum response in the region of 3000 A and a fairly strong response down to about 1600 A. Using such a tube an increase in pulse height by a factor of fourteen has been reported. (ref. 3)

There remains, still, a need to further match the light output to the response of the photomultiplier. This brings up the question of wavelength shifters. There are various substances which have been used; among the common ones are: quaterphenyl, diphenylstilbene, tetraphenylbutadienne and nitrogen. (ref. 1 & 6) W. Lorenz and U.H. Lauterjung (ref. 7) did a series of tests using diphenylstilbene, quaterphenyl, sodium silicate and anthracene. It was found by Lorenz that diphenylstilbene, coated around the entire counting chamber produced both the best resolution and greatest pulse height. Tetraphenylbutdienne is somewhat better than quaterphenyl as well

as diphenylstilbene but has the disadvantage of deteriorating with time. (ref. 2)

Sayres and Wu (ref. 3) report an increase in pulse height, using xenon and a quartz faced photomultiplier, of 1.6 when quaterphenyl is used. An increase of pulse height of 2.5 was reported by Northrup under the same conditions. (ref. 2). Northrup reports a maximum pulse height with a thickness of 20 $\mu gm/cm^2$. The light output remains essentially constant between 20 and 100 $\mu gm/cm^2$.

There is a general preference to use quaterphenyl rather than diphenylstilbene although the author was unable to find the reason for this. Diphenylstilbene produces pulses roughly five-quarters as large as those produced using quaterphenyl and the resolution is better by about 3%. (Roughly 13% compared to 16%)(ref. 7). Some workers claim that nitrogen works well as a wavelength shifter. More will be said about this below.

There is general agreement on the basic construction of the scintillation chamber. Only two workers (ref. 2 & 3) put the photomultiplier inside the counter. All other workers used a quartz window to view the scintillation chamber. The reason for this is that the pressures used were higher than those that the photomultiplier could stand. All chambers were coated on the inside with MgO, which in turn was covered by a wavelength shifter by those who used a wavelength shifter. Shamu (ref. 4) used a polished aluminum chamber which in turn was covered with 1 mm. of MgO . The reason for the aluminizing is that even very thick layers of MgO transmit light. (ref. 10) An MgO-quaterphenyl surface is described as being at least 90% efficient. (ref. 2)

The track lengths of 5 Mev. alpha particles in one atmosphere of xenon, krypton and helium are 2.2, 3.0 and 21.0 cm. respectively. (ref. 3) All chambers were of the order of a few centimeters in length and 5 centimeters in diameter. The chamber used by Engelke (ref. 6)

was in the shape of a truncated cone while that used by Shamu (ref. 4) was hemispherical.

As was mentioned above, nitrogen has been used as a wavelength shifter. Eggler and Huddleston (ref. 1) and Travendale (ref. 5) have reported similar results using nitrogen mixed with helium and argon. .1% N_2 in He and 10% N_2 in A. have produced maximum pulse heights. Engelke claims that at low pressures (less than a few atmospheres), small amounts of nitrogen added to xenon will increase the pulse height (ref. 6).

It is possible that nitrogen will act as a wavelength shifter, but since the excited states of nitrogen are predominantly molecular, rather than atomic, their lifetimes will be longer. With the exception of Northrup (ref. 2) (see below), very little was said about the decay time of the pulses produced and therefore it is possible that these systems were integrating the pulses to produce the results reported. From this author's own work the rise time of the pulses with large quantities of nitrogen (10%) was 400 nsec. as compared to less than 20 nsec. with pure xenon. Northrup and Nobles (ref. 2) suggest that nitrogen could act as a wavelength shifter in those systems in which there is not already an efficient shifter. This theory is supported by Sayres and Wu (ref. 3) who report that using no other shifter the pulse height using nitrogen never exceeded that produced using quaterphenyl.

Wall effect can be reduced to negligible proportions, for work as a neutron counter, if the pressure can be brought up high enough. There is also the advantage that the efficiency of a neutron counter increases with increasing pressure. It is therefore desirable to know what happens to the pulse height and resolution at increased pressures.

Northrup and Nobles (ref. 2) did measurements of pulse heights for various gases and gas mixtures at 100 cm. Hg. They used a hot and cold uranium purifier and talk about effects of 50 parts per million of nitrogen. (The work done by the author indicates that 50 ppm. of contamination is approximately the amount required to observe contaminant effects.) They report a ratio of pulse heights as follows: NaI:Xe:Kr:A:Ne:He = 72:32:16:5:1:10. (100 cm. Hg.)

They report using mixtures of gases and found that xenon mixed with lighter gases gave only slightly greater pulse heights than those produced by the same partial pressures of xenon alone. This is in excellent agreement with the work of Sayres and Wu (ref. 3), although Sayres and Wu did not compare their results with NaI. Shamu, (ref. 4) who went up to very high pressures, also agrees with the low pressure measurements.

Shamu used a calcium purifier and claimed a signal to noise ratio of 500:1 under the following conditions; An RCA 6342A photomultiplier, quaterphenyl and Po alpha particles. From 8 atmospheres to 20 atmospheres the pulse height increased by a factor of two, which would put it at the level of NaI. Using a He-Xe mixture, in which the partial pressure of xenon was constant at 8 atmospheres, the pulse height was greatest at 20 atmosphere total pressure. As helium was added, bringing the total pressure to 160 atmospheres, the pulse height decreased to 25% higher than it was with 8 atmospheres of pure xenon. (ref. 4)

Work done by Engelke (ref. 6) and Lorenz (ref. 7) tends to disagree with the above. Engelke says that above a few atmospheres the pulse height decreases and then remains constant. He also states that small amounts of nitrogen at low pressures can increase the pulse height. The results he obtained at high pressure could be due to small amounts of nitrogen although this has not been established. (Engelke did not use a calcium purifier)

Lorenz used all glass plumbing and a glass calcium purifier operated at 550° C. He showed a decrease in pulse height with increasing pressure, (xenon and diphenylstilbene) and also a steady worsening of the resolution up to 10 atmospheres. It is entirely possible that Lorenz also had small amounts of impurities since he reports a rather rapid poisoning rate. (20% decrease in 100 hours). Work done by Sayres and Wu (ref. 3) indicates that above 4 atmospheres the resolution will remain constant at about 5% and that the pulse height gradually increases with pressure. It is unfortunate that Sayres and Wu did not go to higher pressures and that the present experiment was not equipped to do so at present. Measurements at high pressure need confirmation.

It is thought that gas scintillators are linear with respect to energy regardless of the mass of the particle involved. Sayres and Wu (ref. 3) imply this but Travendale (ref. 5) reports that pulses from U^{235} fission fragments are 50% higher than what is calculated from light particles. He attributes this difference to columnar electron-ion recombination and his work supports this idea. Again, if the pulses were being integrated the disagreement would be resolved.

The most common purifier used calcium. Northrup and Nobles (ref. 2) have used hot uranium (800° C.) followed by cold uranium. This type of purifier, reportedly, can remove hydrogen. Engelke (ref. 6) ran his gas over hot magnesium.

Having purified the gas, the pulse stability depends on the cleanliness of the system. Sayres and Wu (ref. 3), who evacuated to 10^{-6} mm. Hg., reported no change in pulse height for several days. Lorenz (ref. 7), who evacuated to 10^{-5} mm. Hg., reported a drop of 20% in 100 hours. It is worth noting that cooling the system to dry ice temperature restored the pulses to their original height. (See section on results for the lifetime of the present counter).

The decay constants of pure xenon are as follows: (ref. 2) 75 nsec. at 25 cm. Hg., 30 nsec. at 50 cm. Hg. and 14 nsec. at 100 cm. Hg. It is notable that the time constant goes down with increasing pressure. The stopping time of a 5 Mev. alpha particle in one atmosphere of xenon is something less than 3 nsec., assuming that the specific ionization is constant and that the track lengths are 2 cm.

Northrup and Nobles (ref. 2) also experimented with solid xenon, argon and krypton counters and found the pulse height to be about twice that of the gas counter which makes them comparable to NaI pulses. The rise and decay times of these were found to be less than 10 nsec.

As a neutron detector a solid counter would offer the disadvantage of not having the reaction gas mixed with the scintillator. Neither neon nor helium are liquid at liquid air temperatures.

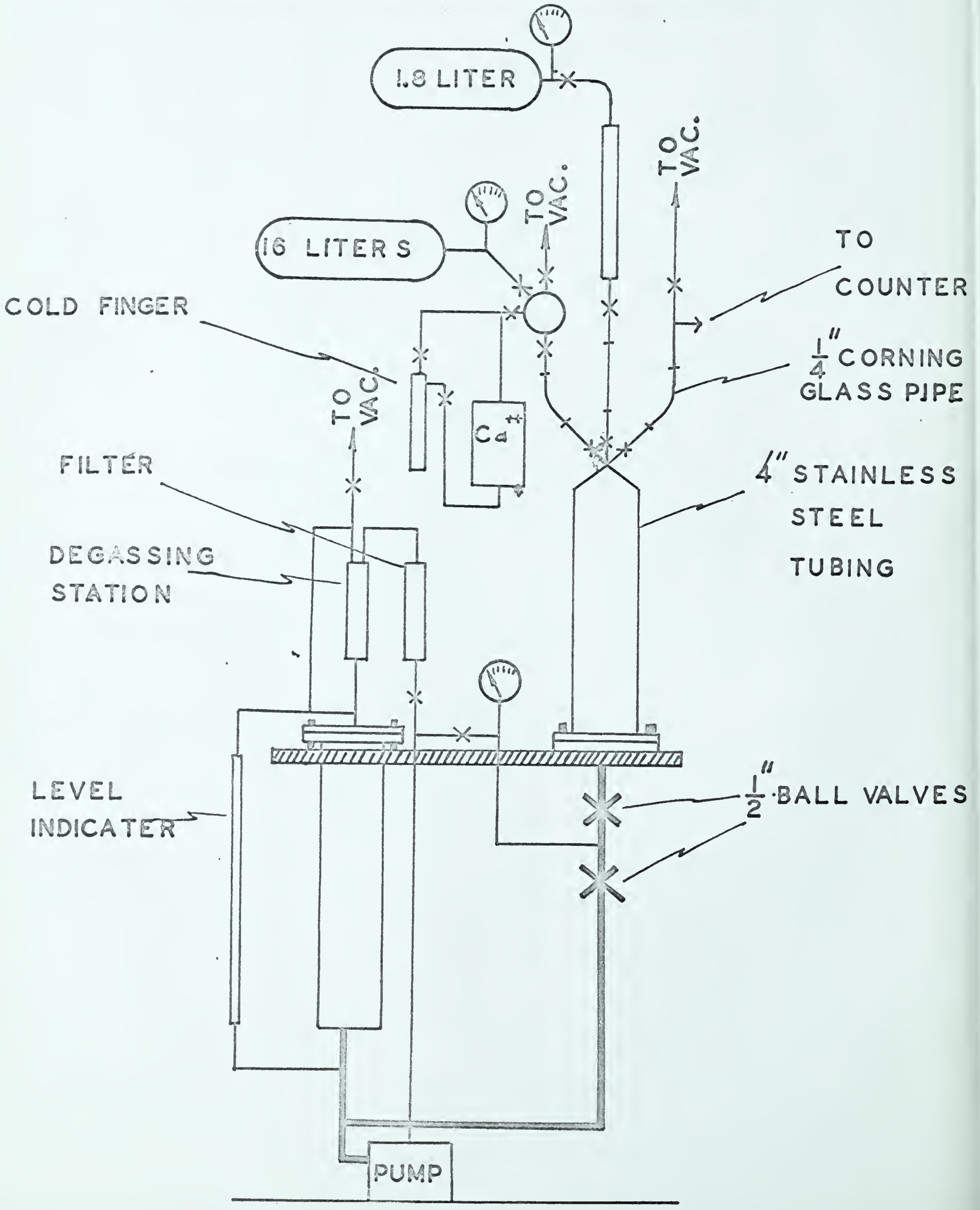
As a neutron detector two reactions have been suggested. These are $He^3(n,p)H^3$, (ref. 2 & 3) and $Ne^{20}(n,a)O^{17}$, (ref. 4 & 6). The cross-section for the He^3 reaction is about 1 barn for neutron energies of 100 Kev. (ref. 12) and has a reaction energy of 770 Kev. The Ne^{20} reaction has a cross-section of about 2 barns at 1 Mev. and a reaction energy of -608 Kev. (ref. 12 & 11). The He^3 reaction would be useful for neutron energies below 600 Kev. The added 770 Kev. of energy would tend to decrease the resolution whereas for energies above about 700 Kev. the subtracted energy of the Ne^{20} reaction would tend to increase the resolution. (See section on predicted behaviour). Sayres and Wu (ref. 3) obtained a signal to noise ratio of 50:1—therefore the noise would correspond to about 100 Kev.

III Equipment

1. Pumping System

In order to facilitate the operation of a counter, a filling system with the following features must be employed:

FIG. 1 PUMPING SYSTEM



a. Storage for gases.

b. Purification of gases.

c. Method of separating gases.

d. A method of moving the gases from one part of the system to another and for filling the counter.

Due to the high cost of He^3, it seemed advisable to build a pump which would recover as much of the gas as possible. Mechanical piston pumps have been used, but this always entailed loss of gas in the valves used to close the bottle.

A fluid piston pump seemed to be the answer to this problem since the fluid could be pumped right into the valve and the valve closed on the fluid instead of the gas.

Fig. 1 shows a semischematic diagram of the pumping system used. The system works for the purpose it was intended but it is somewhat less than ideal. Mercury was proposed for the fluid to be used, but since stainless steel would have had to be used throughout, a silicone fluid was unfortunately chosen. The vapour pressure of the silicone fluid was less than 10^{-3} mm. Hg., but it was messy to work with. Its chief disadvantage is that it dissolves considerable quantities of gas. Experiments were performed to determine the rate at which the outgassed fluid dissolves helium and the result showed that the loss would be tolerable but not negligible. However, for reasons mentioned below, more xenon was lost than was anticipated. Dow Corning "200 fluid"* with a viscosity of 200 centistokes was the first fluid used and proved to be far too viscous, and 10 centistoke fluid was substituted. (Dow Corning "200 fluid"). After almost complete renovation of the system, the 10 centistoke fluid was made to work.

* Supplied by Dow Corning Silicones Ltd., Tipped Rd., Downsview, Metropolitan Toronto, Ontario.

Automatic valves were proposed to control the circulation of the gas through the purifier but since it was thought that convection would circulate the gas, that the gas should not contact the fluid any longer than necessary and that reliable sensing devices could not be made, automatic valves were discarded; however experience shows that there is a need for automatic valves as several accidents have occurred. Human reactions are not quick enough and the fluid has gone too far, which made it necessary to dismantle the apparatus for cleaning.

If mercury were used as a pumping fluid it would offer the following advantages:

1. Mercury has a known vapour pressure and can be removed by a dry ice cold trap, whereas the silicone fluid tended to form a fog.

2. There would be no tendency for mercury to trap bubbles in the body of the liquid. (This has been a bad problem with the silicones)

4. It is very heavy and can be moved through the system rapidly.

5. It would lend itself to the use of automatic valves because, being a conductor, the sensing problem would be eliminated.

Mercury would also offer some disadvantages:

1. It is expensive.

2. It is possible that a pump capable of pumping it at the pressures required may not be found. (A centifugal pump with a radius of 10 inches, turning at 400 rpm. would develop a pressure of 5 atmospheres)

3. It might be hard to keep clean.

4. It has no lubricating properties as with the silicone fluids. Teflon bearings will have to be used.

5. Sudden stoppage of the flow will produce severe hydraulic hammers.

6. Cavitation in the pump may be encountered.

7. It will require all stainless steel construction.

In spite of the disadvantages, mercury appears to be more suitable.

Since it is recommended that a counter using a pressure of 20 to 40 atmospheres be used the pumping system should be capable of handling 300 to 600 psi. It should be pointed out that glass indicators would still be useful on the lower tank since it is never exposed to pressure. It should be easy to keep track of the mercury electrically.

It is worth considering a mechanical pistoned pump with a few hundred cc. of mercury resting on top of the piston. This construction offers no more technical problems than an ordinary mechanical pump and offers all the advantages of a fluid pump. The piston could be powered by connecting it mechanically to an oil hydraulic system of which standard units are available. It is not recommended that both systems share the same cylinder because oil on the cylinder walls would soon contaminate the mercury.

It is not absolutely necessary to build a pumping system capable of handling the pressures used in the counters. Xenon is easily handled by freezing and one could put in the required volume of gas at 0 pressure.* Since 10% helium is adequate the helium could be put in with a low pressure pump after the xenon. This is by far the easiest method but it leaves one in a rather inflexible position.

2. Electronics

At low pressures the track length of the reaction products in the counter is not negligible. The inside of the scintillation chamber was coated with zinc sulfide and pulse shape discrimination was planned in an effort to reduce wall effect. The decay constant of the gas pulses was about 300 nsec. while that of the ZnS was about 10 μsec.

* In order to freeze 1/2 liter of xenon at 40 atmospheres the volume in the cold trap must be at least 20 $cm.^3$

403B AMP. 6688 P.S. 6688 C.F. +250 VOLTS 6688 C.F.

10K 2W
20 T
D=1"
4
1.8K
51K
.1
.1M
82
.1
INPUT
.05
510
33K
390
5-50
1.8K
1W
39K
5.6K 1W
.1
3.9K
75K 1W
.1
27P
82 K
.02
220
33K
47
39K 1W
.1
DR403
3.3K 1W
.1
10K 1W
C1 180P
4.7K
R2
10K
15P
10K
R1 47K
.01
470K
.05
C2 390
8.2K 1W
.1
2.2K 1W
75K 1W
.1
4.7 M
4.7K
DR403
470
47P
20K
47K
47P
470
DR403
4.7K
1K
47PF
+250 VOLTS
.0047
+250VOLTS
VALOR
30C6-4/30
10K 2W
5.6K 1W
39K 1W
.1
.1
.02
22K
27K
82
.1
.1
240K
3.3K .1 GATE
IN903
IN100
15K
.02
8.2K 1W
.05
82K 1W
33K
390
2.2K
.02
.05
.02
470K
10K 10W
47K 1W
240
.1
.05
10K 2W
2.7K 1W
220P
56P
5P
5P
IN100
47K
510
4.7K
15P
47K
1.5K 1W
403B AMP. GATE 6688 C.F. OUTPUT 6688 AMP. 5965 TRIG.

The amplitude of the ZnS pulses was considerably higher than that of the gas pulses and therefore very little energy expended on the wall would produce a long tail on the gas pulse.

Fig. 2 shows the schematic diagram of the circuit required to discriminate the above mentioned long tailed pulses. The input pulse is assumed to be less than 2 volts. The circuit is unstable to larger pulses, but will adequately discriminate pulses as low as .2 volts.

Upon input the pulse is immediately divided, one part is integrated slightly and stored in a 3 μsec. delay line. (Valor type 3006-4/30) The other part is amplified and goes to a phase splitter. The variable capacitor on the cathode of the phase splitter controls the amplitude of the plate signal by supplying a variable surge current to charge the distributed capacity on the plate. The anode pulses are differentiated, resulting in an amplitude loss of 50%. Another cathode follower is required so that sufficient current can be supplied to the pulse stretching circuit.

The diode and the network of capacitors and resistors after V_3 shape the pulse to conform exactly to the incoming pulse. There is a slight undershoot on the incoming pulse. (.05 volts in about 5 volts) This undershoot was enough to completely cripple the operation of the circuit. C_1,C_2, R_1 and R_2were introduced to match the undershoot of the manufactured pulse to that of the incoming pulse.

Since the shape of the manufactured pulse is very sensitive to the load another cathode follower is required.

The two pulses do not have the same rise time and their peaks are not exactly alike making direct addition of the pulses impossible. Therefore the pulses are clipped before adding so that only the tails are compared. A.C. coupling of the adding network introduced undershoot problems again.

To remedy the A.C. coupling problems the whole ciruit was D.C. coupled. The bias of the cathode follower is controlled by the phase splitter. The 50 μamp. current flowing along the bias line biases the cathode follower 2 volts higher than the phase splitter. This 2 volt difference is used to bias the clipping diodes and a balance is established using the 20K pot.

If there is output from the adder it is amplified by V_7 and sent to the trigger circuit V_8.

The incoming pulse which was put into the delay line is amplified before it goes to the gate. The gain of the amplifier is about 14, the attenuation in the delay line is about 2.9. The overall gain from input to output is therefore about 5.

The gate is a swamping type. The 1N903 is normally back biased and if there is no trigger pulse, it will remain that way upon the arrival of the pulse from the amplifier and the pulse will get to the output cathode follower. If there is a gate pulse, then approximately 6 MA. is flowing through the diodes (Gate pulse is approximately 40 volts) and the incoming pulse is swamped. (The drop across the 1N100 is held constant at -.4 volts.) The advantage of this type of gate is that it is linear, but it does have the disadvantage of having the negative step.

The circuit is very sensitive. If the tail on the incoming pulse is .04 volts or higher it will cause the ciruit to trigger. With an input pulse of the maximum 2 volts, this amounts to 2% by amplitude.

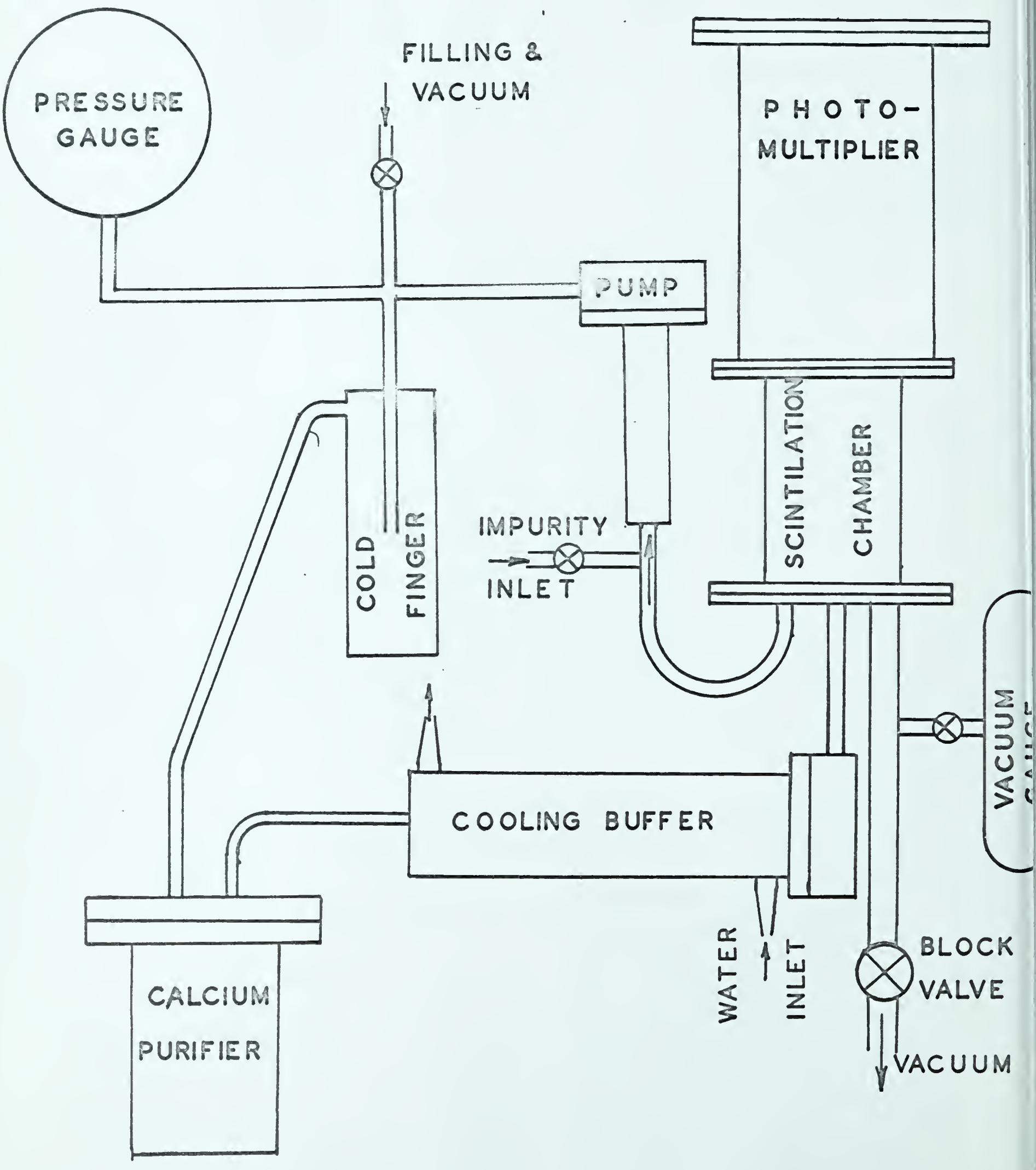
3. Counter system

a. General arrangement

Fig. 3 shows the arrangement of the component parts of the system. Although it was hoped that convection would circulate the gas a small pump was built to insure circulation. During the impurity measurements, the purifier was cold and the only way to circulate the gas was by means

FIG. 3 COUNTER SYSTEM

SEMISCHEMATIC - NOT TO SCALE



of the small pump. The pump consisted of a ball bearing valve in the head and an unlubricated steel piston closely fitting a brass sleeve. The piston was moved by a solenoid which was around the body of the pump and the solenoid in turn was alternately turned off and on by a commercial flasher. The piston had a hole drilled in it and a teflon plug acted as a valve. The pump circulates gas at approximately 2 cc. per sec.

The gas was passed through a cold buffer before passing it into the counting chamber. The buffer consists of a stainless steel tube with a friction seal (described in the section on the purifier) and cold water jacket. A stainless steel slug was placed in the tube to force the gas against the walls of the tube. The back of the slug (next to the inlet from the purifier) was stained by evaporating molecules from the trap which proves that the buffer is required.

The calcium purifier and the scintillation chamber are described in their own sections. The large block valve on the bottom of the scintillation chamber was required to obtain sufficient pumping speed. The whole system was outgassed for 24 hours at 150° C. Further details are given in the section on results.

b. Purifier

As was stated previously, the gas purity is of utmost importance and the purification proved to be the most difficult problem. Calcium was used as the purifying agent.

The first purifier was one which would go inside the counting tube for compactness. This idea failed although considerable work was put into it because it was not possible to insulate the purifier from the rest of the system. (A sodium getter could be used once the gas is purified)

It was decided that an external purifier was required and the first one was made of brass with an aluminum compression seal. It was operated at 150° C, and as this purifier showed no signs of working it was decided that the temperature should be raised to 400°C. To stand the higher temperature the purifier was rebuilt out of stainless steel with an aluminum compression gasket. Unfortunately the compression seal would not stand up to the temperature variations and leaked very badly. The purifier was again rebuilt using a "Friction seal" as suggested by reference 13. This seal is basically a tapered plug fitting into a matching hole between which there is a soft metal. The gasket is easily formed by cutting a ring out of shimstock and bolting it between the top and the bottom pieces. Using 1/4" flanges and six 10-32 bolts a helium leak tight seal, which would remain sealed through all temperature variations, was easily established.

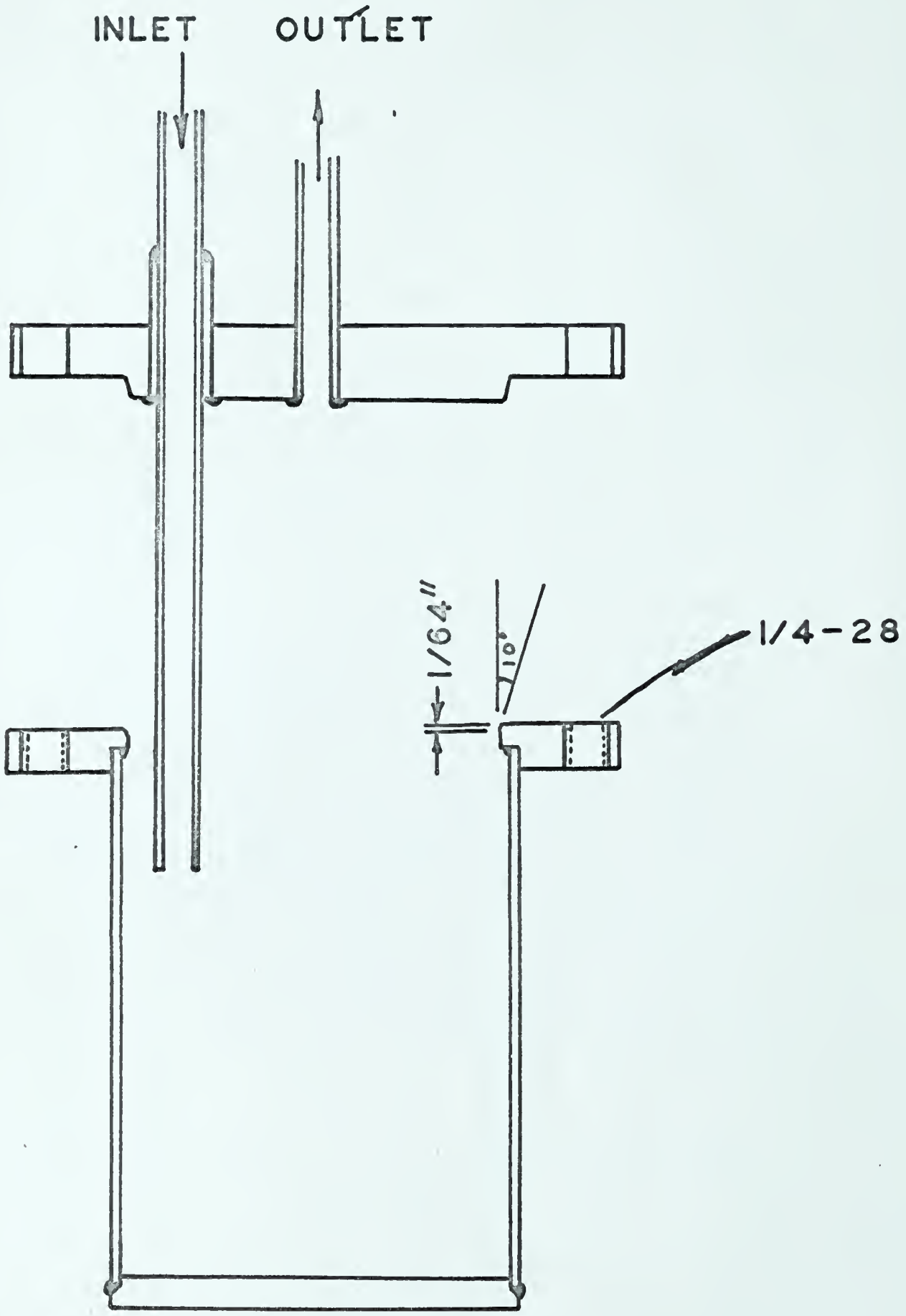
The purifier was heated to 400°C and the whole system outgassed for two days. Xenon was then introduced. The system showed no signs of working and was consequently dismantled. The inside of the counter chamber was blackened, presumably from the purifier. The pipe leading from the purifier was cooled with water but it was obvious that something better was required. The cooling unit, shown in Fig. 3 was built.

Silver solder would not stand up to the action of the calcium at 400°C. The trap was rebuilt again, and all joints were argon arc welded. Copper was again used as a gasket. The new system was outgassed for 3 days, and after having filled the counter, gas pulses were barely visible over the noise of the photomultiplier.

It was at this time that grave doubts were being raised regarding the purity of the gas. A mass spectrometer analysis showed that the gas was .07% nitrogen, with lesser amounts of CO_2 and water. (Later

FIG. 4 CALCIUM PURIFIER

SCALE = FULL SIZE



MATERIAL — STAINLESS STEEL

ALL JOINTS — HELIUM ARC WELDED

GASKET — NICKEL

TEMPERATURE = 450° C

results show that this amount of nitrogen would bring the pulse height down by a factor of two.)

Following the recommendations of Sayres and Wu (ref. 3), the temperature of the purifier was raised to 500° C during the outgassing. It was only a matter of hours before the copper gasket had been eaten entirely away by the calcium.

Copper was obviously unsuitable as gasket material and many metals were tried before using nickel. (Nickel is used in commercial calcium purifiers) Much difficulty was experienced in getting nickel to seal. Many variations of the sealing surfaces on the trap were tried. 1/4 - 28 bolts were substituted for the 10-32 bolts used before. The flanges bent before a seal was effected even with a sealing surface only 1/64" wide.

It was suggested that the trap should be assembled while the top and bottom were at different temperatures. The top was thoroughly soaked in liquid air and then the body of the purifier was bolted on as quickly as possible and as tightly as the flanges would allow without bending. A preformed gasket was used.

The calculated change in diameter at the sealing surfaces was .008" which gives a .004" change for each side. The gasket was .006" thick and a reliable seal was produced.

The purifier shown in Fig. 4 is the final model used. A nickel gasket was used and the purifier was heated to 550° C for outgassing and 450° C for normal use. At this temperature (450°C) the purifier could remove gross amounts of nitrogen almost as fast as the circulating pump could pump it in. It should be pointed out that the body of the trap was somewhat hotter than the top, where the thermocouple was located.

In the future all calcium traps should be made along the same lines

as described above but with heavier flanges (at least 3/4" thick stainless steel) and heavier bolts. Provision should be made for gripping the top of the purifier so that the bolts may be turned. The purifier should be made entirely of stainless steel, and all joints should be stainless steel helium arc welded. The gasket should be nickel and the sealing surface should be about .030" wide. The running temperature should be at least 450°C.

The trap shown in Fig. 4 was surrounded by an asbestos box with 1/2" wall thickness. The heating coil was wound around a piece of stainless steel tubing, with asbestos insulation, mounted on the bottom of the insulating box. The thermocouple (Cromel-Alumel) was placed between the top of the trap and the top asbestos plate. When assembled, the body of the trap was contained within the heating coil. Approximately 185 watts was required to heat the purifier sufficiently to get a thermocouple reading of 500°C. The insulation box was disassembled while the power was on and it was observed that the body of the trap was dull red hot. (It required subdued light to see it which places the temperature at approximately 700°C)

When cleaning the purifier, one must be careful. Calcium nitride is highly reactive to water and will form ammonia. After initial cleaning the final bits and pieces can be removed by soaking the purifier in nitric acid. Disassembling the purifier is a problem; the top is stuck rather firmly onto the bottom. Grub screw holes may be put into the top and through these screws may be turned to force the bottom off.

Unfortunately, the successful construction of a calcium purifier does not end the purification problem. Any hydrogen in the system may combine with the calcium nitride to form ammonia. Ammonia will

not be removed by calcium and activated charcoal was placed in the cooling tube in the hope that it would absorb any ammonia that would form. (It is thought that ammonia was not present in the system, initially, because of the repeatability of the results.) It is not clear that the charcoal was doing what it was intended to do. Freezing the ammonia will remove only gross amounts. The vapour pressure of ammonia at dry ice temperature is 40 mm Hg. and at the boiling point of xenon it is 1 mm. Hg. There is a short discussion in the results section on this subject. 10% nitrogen was introduced and sufficient hydrogen could have entered to form significant amounts of ammonia.

c. Scintillation chamber

The rise time of zinc sulfide pulses with Pu alpha particles is about 400 nsec. and the decay constant is about 10 μsec. Zinc sulfide pulses are roughly 10 times higher than corresponding pulses from plastic phosphor.

The photomultiplier was initially put into the counting tube so that there would be no problem with windows. The first scintillation chamber tried was 5 1/2" long and 1 3/4" in diameter. It had a zinc sulfide coating on the inside and there was a removable Pu source at the end.

Coating the chamber with zinc sulfide was a problem. Sodium silicate was used as a binder because there is no hydrogen in it to produce background for use in a neutron counter. It was later discovered that sodium silicate hydrates when it hardens. The counting tube was turned in a lathe and the sodium silicate was spread in it. To obtain a uniformly thick coating, a small amount of detergent was mixed with the sodium silicate. The sodium silicate could have been dissolved

FIG. 5B LIGHT SLEEVE

SCALE = FULL SIZE

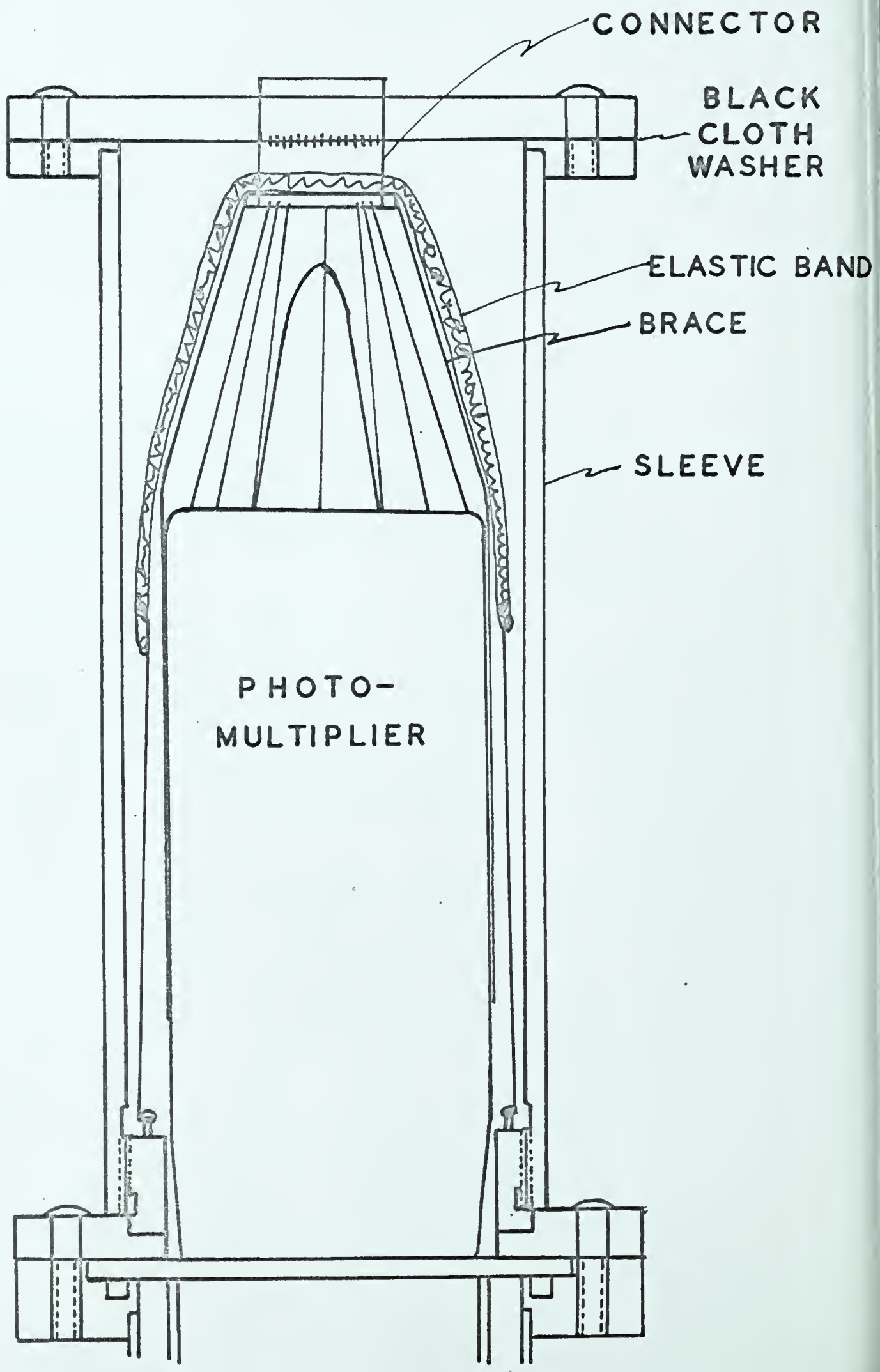
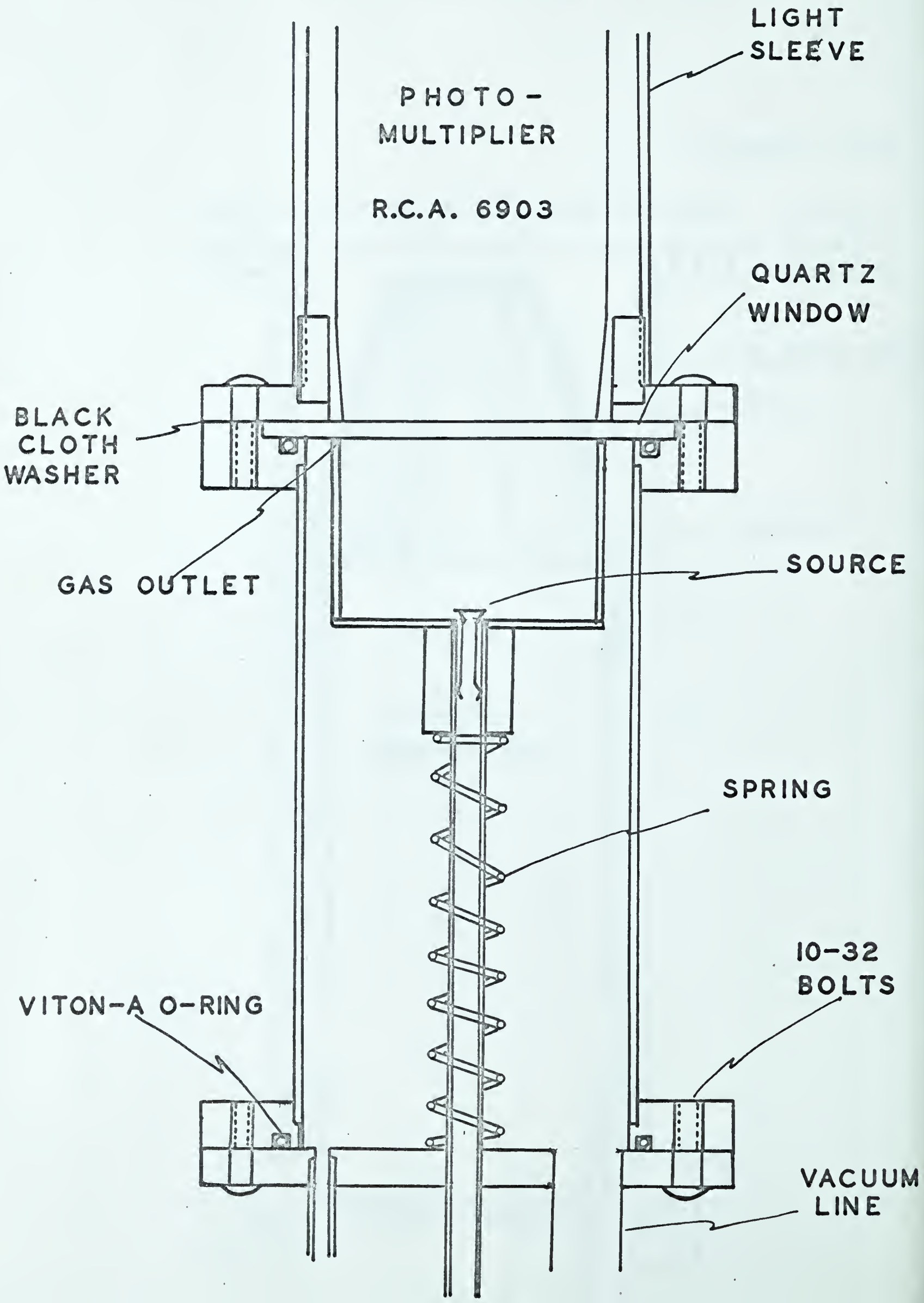


FIG.5 SCINTILLATION CHAMBER

SCALE = FULL SIZE



in alcohol and sprayed on to accomplish the same result. (ref. 7) The zinc sulfide was powdered and a powder atomizer was built. This device consisted of a florence flask in which the powder was agitated by blowing high pressure air into it through a narrow nozzle. The air came out of a wide tube carrying the powder with it.

As was mentioned in the section on the purifier, barely detectable pulses were seen using a 400° C trap. It was the above described scintillation chamber which was being used at that time. This was replaced by a similar MgO coated chamber. There was no change in the behaviour of the counter. Gas pulses were barely discernable over the noise. The gas pressure was about 1/2 atmosphere. Quaterphenyl was coated in each case on the photomultiplier tube face only, because of difficulties in evaporation it was not possible to gat it into the scintillation chamber.

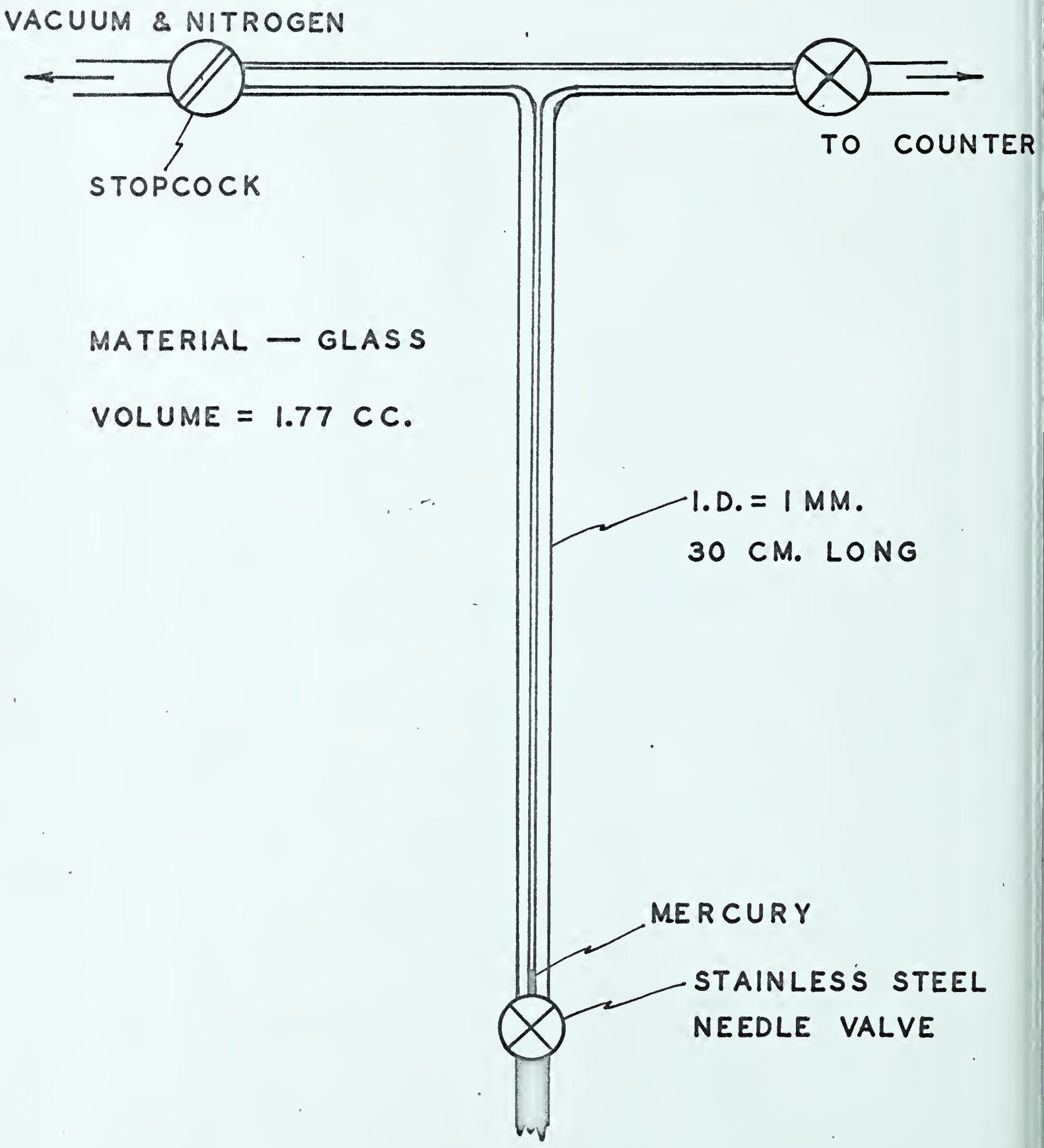
It was not until a short scintillation chamber was constructed (1 7/8" in diameter and 1 1/4" long) that definite results were obtained. At this time the photomultiplier was also moved outside the chamber so that the whole system could be outgassed. (The photomultiplier tube cathode will not stand up to heat.) Fig. 5 shows the final counter used.

The inside of the system was not coated with quaterphenyl because the impurity measurements required only relative results. Quaterphenyl has been quoted as having contaminant effects and it would not have stood up to the outgassing treatment. The photomultiplier had a quaterphenyl coating.

The system was outgassed for a day at 150° C with the calcium trap at 550°C. Viton-A O-rings were used throughout the system but they did not live up to their advertised characteristics. They became deformed, but since they remained helium leak tight it was decided to to continue using them. The O-ring valves used on the system leaked

FIG. 6 IMPURITY INJECTOR

NOT TO SCALE



when they were moved. After outgassing the system the valves were closed off and the pressure remained at 10^{-4} mm. Hg. for several hours without moving. (Trap temperature was 450° C) After cooling the trap to room temperature the pressure dropped, valves still closed, to 10^{-5} mm. Hg. and stayed there for several hours without changing.

Xenon was put into the system and frozen in the cold trap. The excess air was pumped out by a roughing pump and the gas was allowed to warm and flow through the calcium trap. (455°C) The final pressure was 53 psig. and gas pulses were seen immediately. It took only a few minutes to reach maximum pulse height.

The second time this system was tried, it was outgassed for only 4 hours at 150°C. The pressure was only 2 times 10^{-4} mm. Hg. with the purifier hot but it remained at this level and therefore xenon was put in. The final pressure was 55.5 psig. Again almost immediate results were obtained.

The photomultiplier had a coating of quaterphenyl and was light coupled to the quartz window with silicone fluid.(500,000 cs.) The silicone fluid did not affect the quaterphenyl coating. The best signal to noise ratio was obtained with a tube voltage of 900 volts. This is discussed in the section on results.

d. Impurity Injector

A device, which would inject controlled amounts of impurity gas, against the pressure in the counter, is shown in Fig. 6. Mercury is filled in the small bore tube which is 1 mm. in diameter. 1 cm. movement of the mercury in this tube corresponds to a volume change in the device of 8 times 10^{-3} cm^3.

To use it the entire system was allowed to assume room temperature. Dry nitrogen was put into the injector to a pressure just below the pressure in the counter. The valve into the counter was opened briefly

and some of the xenon flowed into the injector, thus equalizing the pressures. The gases were allowed to diffuse for 15 minutes. Then the valve into the counter was again opened and the mercury was moved up the required amount to inject the desired volume of nitrogen.

The volume of the counter was .8 liters, thus 1 cm. of movement in the small bored tube would put 10 parts per million of the gas mixture into the counter. It is clear that a better method is required if further and more precise impurity measurements are desired. The error in this method could be as much as 30%.

IV Predicted behaviour of a neutron counter.

In this section an attempt will be made to predict the behaviour of a gas-neutron counter and spectrometer. Both the He^3 and Ne^{20} reactions will be considered. The behaviour of the scintillator will be assumed to be optimum.

It will be assumed that the pressure in the counter will be 20 atmospheres of xenon and 4 atmospheres of the reaction gas. The track length of 5 Mev. alpha particles in this much gas is only 1mm. and that of protons and tritons is roughly twice that for the same energy. Wall effect is considered to be negligible.

Consider first the lifetime of the counter. The present counter was outgassed for 4 hours at 150°C. It did not have quaterphenyl inside and had previously been more thoroughly outgassed. It can only be said that the lifetime of the present counter was in excess of one week. Sayre and Wu (ref. 3) state that the presence of quaterphenyl does not measurably shorten the life of the counter. They report a lifetime of a few days.

The author lowered the temperature of the photomultiplier tube and scintillation chamber with dry ice and the pulse height was raised by about 10%, but the chief advantage of lowering the temperature

FIG. 7 RESOLUTION

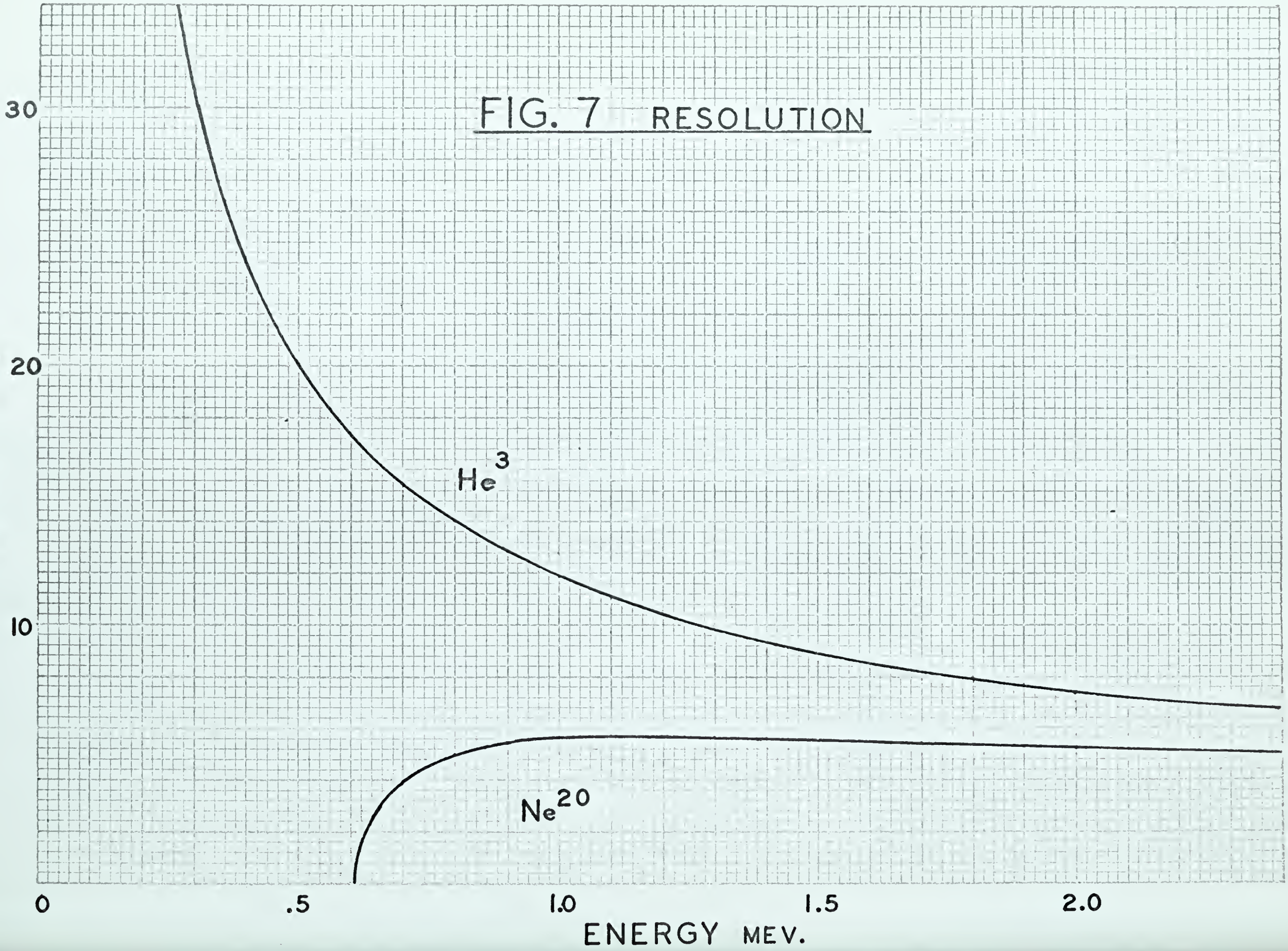


FIG. 8 EFFICIENCY H_o^3

EFFICIENCY X 10^4

100

10

1

.01

.1

1

10

ENERGY MEV.

of the entire counter is that the vapour pressure of the quaterphenyl will be lowered. It is predicted that a lifetime in excess of a week should be easily realized.

Using 5 Mev. alpha particles the author was able, after prolonged and continuous use of the photomultiplier tube, to obtain a signal to noise ratio of 40:1. This was not significantly improved by lowering the temperature. The use of quaterphenyl in the scintillation chamber may raise the light output by a factor of 2.5. (ref. 2)
Raising the pressure of the xenon to 20 atmospheres should raise the light output by a further factor of 2 (ref. 4) resulting in the naive prediction of a signal to noise ratio of 200:1. It would not be unreasonable to expect a signal to noise ratio of 100:1 using 5 Mev. alpha particles.

Sayres and Wu report a resolution of "around 4%" (ref. 3) Their resolution improves with pressure and it will be assumed, for the purpose of calculation, that a resolution of 4% may be obtained at 20 atmospheres. (Pu alpha particles.)

Consider now what will happen in a neutron counter using He^3 when a 1 Mev. neutron reacts in it. The energy dissipated in the counter is 1.77 Mev. It will be assumed that the resolution varies inversely with the square root of the energy. 1.77 Mev. could therefore be measured with a resolution of about 7%. Subtracting the reaction energy the neutron energy can therefore be measured with a resolution of 12%. Graphs of the predicted resolutions, under the best conditions, are shown in Fig. 7.

The efficiency will be roughly the same for both counters(helium or neon) The reaction cross-section for the $He^3(n,p)H^3$ reaction increases at the low energies.

The efficiency has been computed using the cross-sections as reported in reference 12 and has been plotted against energy in Fig. 8.

It was assumed that the counting chamber was cylindrical and that it was 1 1/4" Long.

There will be background caused by neutron collisions on the He^3 nuclei. The maximum energy transfer in such a collision is 3/4 of the neutron energy. Since 770 Kev. will be subtracted off the energy, any collisions of neutrons with less than 1 Mev. of energy will not cause any difficulty. Spectra involving neutron energies above 1 Mev. would have to be corrected. Using the neon reaction one would encounter this problem with the low energy neutrons since the Q of the reaction is negative. Maximum energy transfer in this case is .23 of the neutron energy.

V Results

The purifier was outgassed at 550° C. and the system was outgassed for 24 hours at 150° C. before xenon was put in. The xenon was frozen and pumped on and then allowed to warm and flow through the purifier into the counting chamber. The final pulse height was attained almost immediately. The system was partly dismantled and air was allowed into the system for a few hours. The system was then outgassed for 4 hours at 150° C. Again the final pulse height was attained almost immediately.

The system as described in earlier sections and outgassed as above was stable for at least a week.. Time would not permit a further life test.

The results for low level impurity measurements are shown in Fig. 9A & 9B and pulse height against pressure in Fig. 10. The rise time of the pulses was less than 20 nsec. The decay constant was about 300 nsec. After each test the gas was purified. In each case the pulse height returned to the original value. The pressure was taken from high to low so that uncertainties regarding composition were avoided.

At the low pressure (5psi.), .1% nitrogen was introduced. This

brought the pulse height down from .4 volts to .2 volts. The purifier was heated (450° C) and then 10% nitrogen was introduced. This brought the pulse height down from .4 volts to .05 volts, and increased the rise time to 400 nsec. The purifier took 4 minutes to remove one half of the nitrogen. Calculation shows that this is approximately the time required to circulate roughly 1/2 of the gas through the purifier. It required another 4 minutes to remove another 1/2 of the remaining nitrogen. At this time the pulse height was .1 volts.

It is impossible to say beyond this how much nitrogen was present at a given time but the pulse height rose rapidly. In eleven minutes from initial contamination the pulse height was .15 volts and the rise time had returned to 20 nsec. From the initial introduction of the nitrogen it required 25 minutes to bring the pulse height up to .3 volts and an additional 1/2 hour to bring it up to .35 volts. The pulse height never rose above that voltage on the purifier alone.

If one lowered the pressure, by freezing some of the xenon in the cold trap, the pulse height would again rise and if one lowered the temperature of the photomultiplier and scintillation chamber with dry ice, the pulse height would rise 10%. It should be noted that prior to the introduction of gross amounts of nitrogen the pulse height was constant at .4 volts from the lowest pressure to about 10 psi. It is thought that enough hydrogen was introduced with the nitrogen to produce significant quantities of ammonia. (See discussion)

Tests done on the photomultiplier show that the signal to noise ratio increases steadily as the tube voltage decreases. However one gains in signal to noise ratio at the expense of resolution. On the counter no significant change in signal to noise ratio was observed below 900 volts and so all tests reported were done at 900 volts. The signal to noise ratio at this voltage was 40:1.

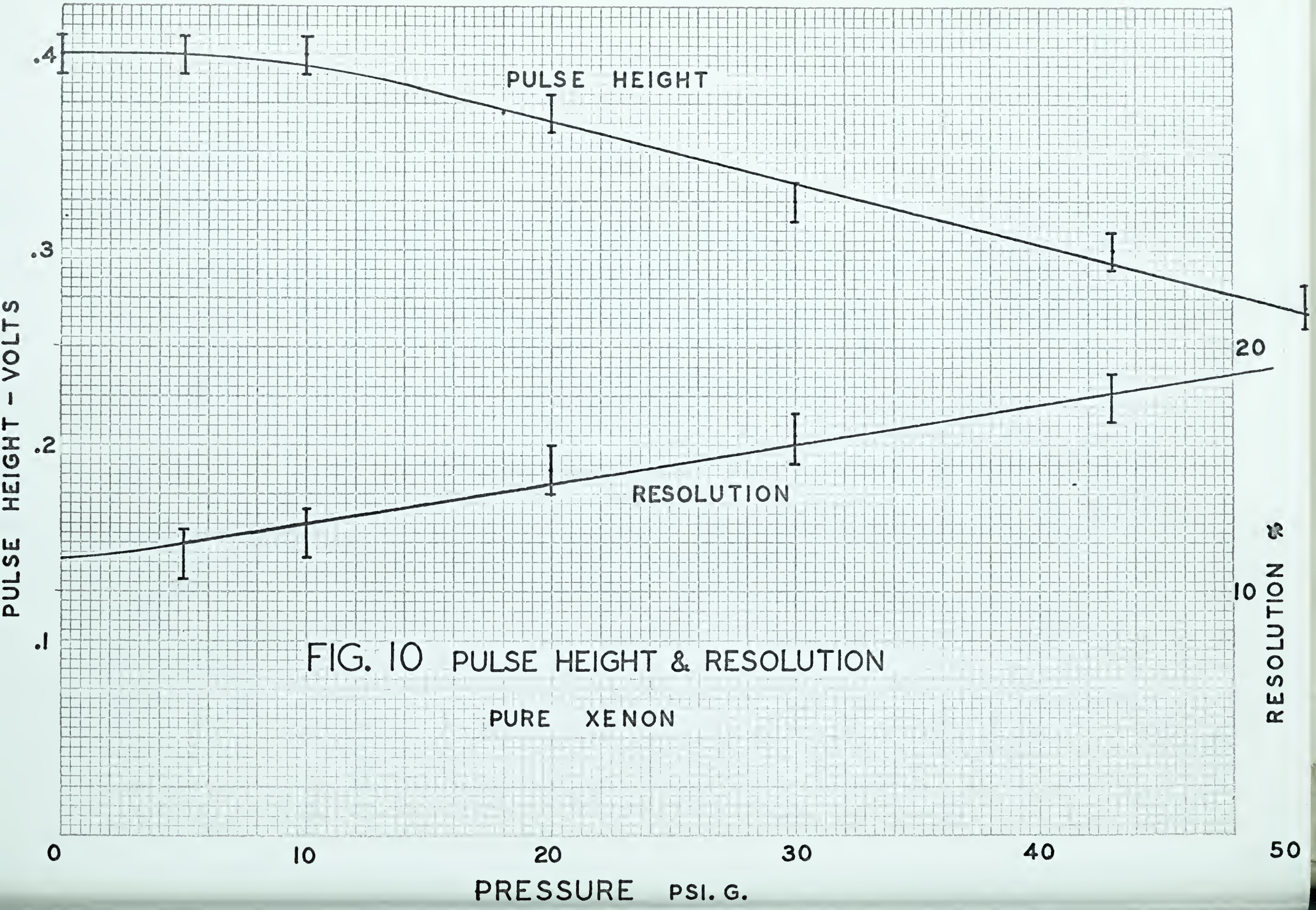


FIG. 10 PULSE HEIGHT & RESOLUTION

PURE XENON

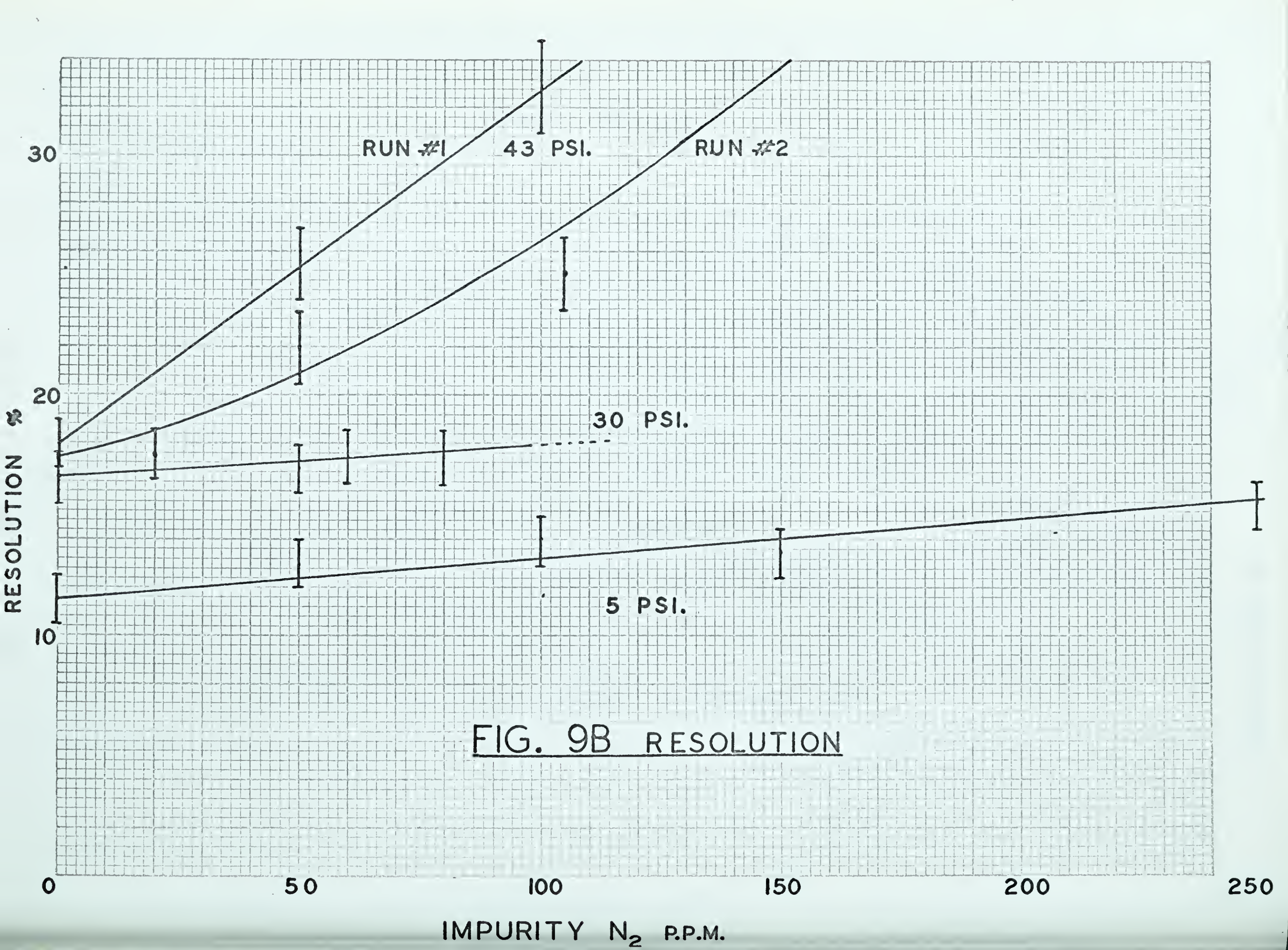


FIG. 9B RESOLUTION

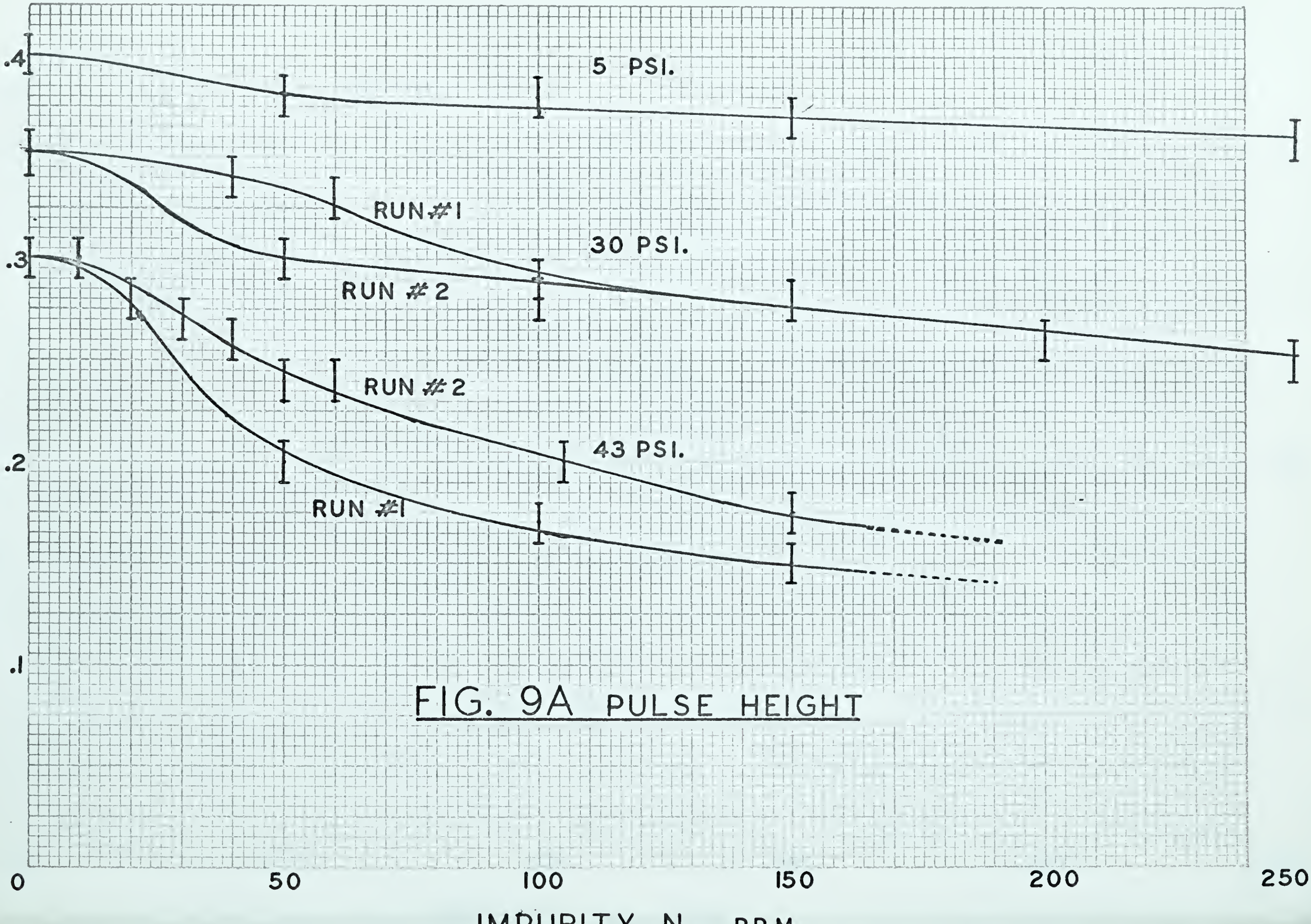


FIG. 9A PULSE HEIGHT

VI Discussion

The most significant trend which comes out of the impurity measurements is that the lower the pressure the less sensitive is the gas to a given percentage of impurity. However, it must also be remembered that at 45 psi. there is 4 times as much nitrogen present for a given percentage impurity as there is at 0 psi. It would seem that the effects of impurities should be measured in terms of the partial pressure of the contaminant rather than in terms of the percentage.

Two runs were done at the high pressure. They do not agree, and this is attributed to inaccuracies in measuring the true contaminant. No effect was noticed at 10 ppm. At this pressure 10 ppm corresponds to a partial pressure of roughly 30 microns.

The results of these tests tend to form a bridge between disagreements outlined in the literature. It disagrees with Engelke (ref. 6), who claims that nitrogen, at low pressures, will increase the pulse height, but it should be noted that at low pressure the decrease in pulse height is not as pronounced.

The resolution and pulse height both improve with decreasing pressure, which is in disagreement with Sayres and Wu (ref. 3) and Shamu (ref. 4); however this is attributable to the type of chamber used. No quaterphenyl was used inside and the source was rather large and dark. At the high pressures the tracks will be confined to a region close to the source and light reflection would not be efficient there. At the lower pressures the tracks could lengthen into a more efficient region.

For work as a neutron counter a source would not be required. A source of thermal neutrons would be required for calibration purposes. The counting chamber would have to have a hole in it somewhere, to allow admittance and circulation of the gas, but it should not cause difficulties.

A neutron counter could now be built without further testing except for the problem of the ammonia formation. The introduction of 10% of nitrogen introduced 50 ppm. of hydrogen because the nitrogen used contained this much. There is also the probability that some water vapour, and oil vapour entered the system with the nitrogen. It is the opinion of the author that it is very likely that several hundred parts per million of hydrogen entered by the above mechanism. It is estimated that roughly 50 ppm. of ammonia would be required to produce the effects noted. There is definitely an impurity in the gas which will not be removed by the calcium, nor will it be removed by dry ice.

Whatever the impurity is, it can be removed by freezing it with liquid air. This process unfortunately freezes the xenon as well but in thawing, the xenon arrives back at the counting chamber before the impurity because full pulse height is observed for about 10 minutes after warming.

It is the opinion of the author that ammonia will present a problem although it has still to be confirmed that the impurity is indeed ammonia. Charcoal should remove it and it is possible that the charcoal used in the system became saturated.

The new counter should be able to go to 20 atmospheres pressure. It should be thoroughly outgassed and should have a coating of quaterphenyl around the inside, including the inside face of the window. Provision should be made to keep the counter at dry ice temperature. The problem of getting quaterphenyl into a thoroughly outgassed system is discussed below. Viton-A O-rings may still be used on the system but it is recommended that teflon or metal gaskets be used.

The counter itself should not require a calcium trap on it since the stability of the system has been found to be sufficient as not to require continuous purification. It will require a cold trap and purification of the gas will be required as the counter is being filled. Under no circumstance should the counter be allowed to come into contact with

any amount of contaminant. It was found that the brief contact with 10% of nitrogen was sufficient to cause poisoning within a week and purification was required.

It is recommended that two cold traps and a purifier be employed and outgassed with the system. The first cold trap would come before the purifier, and the second would be on the counter. The calcium purifier should be accompanied by a charcoal trap.

Quaterphenyl will not stand up to the outgassing treatment. Provision should be made to remove the quartz face in a vacuum under a bell jar, evaporate the quaterphenyl, and reassemble it without the inside of the counter's having been in contact with air. Air pressure will keep the quartz pressed against its gasket and it will be necessary to handle the window only and not the retaining ring required for high positive pressures.

The evaporation of quaterphenyl is extremely tricky. Close control would have to be implemented if the above procedure is followed, so that a known thickness is sure to be deposited.

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